

## Molecular Dynamics Simulations of Iron-Joining Using Copper as a Filler Metal

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### Abstract

The study of the liquid filler metal infiltration on the narrow channel of adjoining metal bears importance in understanding the mechanism of the metal brazing process. In this study, we employed the molecular dynamics simulation to understand the mechanism of Cu liquid infiltration through the narrow channel of Fe slabs. Our simulation showed that the wetting process of Fe surfaces by Cu liquid precedes the infiltration process. This study also revealed that the channel became narrower and blockages were found in the channel due to the deformation of Fe surface. In addition to the effect of viscous drag, this process should also contribute to the decreasing speed of the Cu liquid front.

### Abstrak

**Simulasi Dinamika Molekular Proses Penyambungan Besi menggunakan Tembaga sebagai Logam Pengisi.** Studi mengenai infiltrasi logam pengisi pada celah sempit sambungan logam sangat penting untuk memahami mekanisme proses pematiran logam. Pada penelitian ini kami menggunakan simulasi dinamika molekular untuk memahami mekanisme infiltrasi cairan Cu melalui saluran sempit Fe. Hasil simulasi menunjukkan bahwa proses pembasahan permukaan Fe oleh cairan Cu mendahului proses infiltrasi. Diketahui pula bahwa saluran menjadi lebih sempit dan ditemukan penyumbatan pada saluran akibat adanya deformasi permukaan Fe. Selain efek aliran kental, proses ini juga berkontribusi terhadap penurunan kecepatan cairan Cu.

*Keywords: metal brazing; Fe; Cu liquid; infiltration; molecular dynamics simulation*

### Introduction

Two pieces of metals can be joined by infiltrating a filler metal into the joint. This method is called brazing. In brazing, two pieces of metals can be joined by infiltrating a filler metal into the joint. The study of filler metal-adjoining metal interaction and the infiltration process of filler metal into the channel between adjoining metals are important in mechanical engineering.

Previous studies have shown that resistance to the fluid flow increases with the length of infiltration due to viscous drag. Therefore, the velocity of the liquid front should decrease with increasing infiltration channel [1-3]. However, these studies failed to verify the effect of the dissolution of adjoining metal and changes in the channel dimension during the brazing process.

Ambrose et al. have performed more specific studies on the liquid Ni - P braze through the narrow channel of

Fe-Cr workpieces [4]. Studies have reported that the first and last stages of the interfacial interaction are relatively stable, whereas the intermediate stage exhibits high reactivity. In other studies, Semlak and Rhines [5] observed the parabolic dependence of the rate for Cu through the Fe channel and Pb through the Cu channel. Asthana [6] suggested that the dynamics of wetting play an important role in the infiltration processes. Several techniques have been proposed to improve the strength of the joining structure. Cao et. al. [7] discovered that the Cu heat-affected Cu zone featured higher tensile load than the Cu interface and Ti interface during the cold metal transfer welding – brazing of titanium to copper. In the study conducted by Zhang et. al. [8], a high welding efficiency can be achieved by using tungsten/metal gas – suspended arc welding. Zhang et al. has proposed a new technique of brazing [9] to improve the strength of brazing joint of sapphire microwave window with oxygen-free copper. They observed that the shear strength of brazing joint

specimens obtained using their technique can reach up to 145 MPa. Difficulty arises from in-situ observation of the infiltration process of filler metals due to the rapid penetration rates and small channel dimensions. Fortunately, these issues can be accessed by molecular dynamics (MD) simulation. Recently, Webb III and Hoyt [10] carried out atomistic simulation to study the liquid Cu metal infiltration through the crystalline Ni channel. Jiao et al. [11] investigated the effect of surface roughness and pore on the linear friction of welding. They observed that surfaces roughness and pore influence the final structure of the weld. However, although the studies on liquids infiltration through the narrow channel have been published, to the best of our knowledge, the number of atomistic simulations for this process is limited. In this paper, we investigate the Cu liquids infiltration through the Fe channel using MD simulation. This study aimed to elucidate the mechanism of the infiltration process at the atomic level. In this study, we observed that the channel became narrower, and a blockage was found in the channel due to the deformation of the Fe surface. This process should also contribute to the decreasing speed of the Cu liquid front.

## Simulation Method

In this simulation, we employed the embedded-atom-method (EAM) potential [12,13] which was developed by Bonny et.al. [14], for Fe and Cu the parameters are provided in the NIST Interatomic Potentials Repository system [15]. We have performed the preliminary calculations to evaluate the performance of this potential [16]. Our calculation results have shown the good agreement with the findings of previous reports on Fe and Cu melting points. The total energy  $U$  - in the EAM potential is expressed by the following equation:

$$U = \frac{1}{2} \sum_{i,j (j \neq i)} V(r_{ij}) + \sum_i F(\rho_i), \quad (1)$$

where the first term refer to the sum of pair potential interaction  $V(r_{ij})$  between atom  $i$  and  $j$ . The second term represents the sum of embedding energy  $F$  of atom  $i$  into the atomic electron density  $\rho_i$  induced by all other atoms. The atomic electron density of atom  $i$  is obtained from the sum of electron densities of neighboring atoms  $j$ :

$$\rho_i = \sum_{j \neq i} \rho_j(r_{ij}). \quad (2)$$

The large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [17] was used to carry out MD simulations. The pressure and temperature of the system were controlled using NPT ensemble with the Nosé – Hoover scheme [18,19]. Each step in the

integration of the equation of motion corresponds to 1 fs. The total simulation time for each process is 180 ps, which is equal to 180000 MD steps.

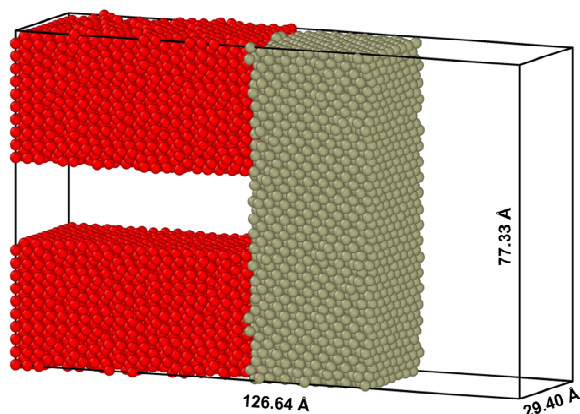
The initial configuration of the simulation system has been constructed. Thus, the infiltration occurs along the  $x$ -direction. The simulation cell measured 126.64, 29.40, and 77.33 Å in the  $x$ -,  $y$ -, and  $z$ -directions, respectively. Two slabs consisting of 8000 Fe atoms were placed in the upper and lower parts, which were separated by a 20 Å narrow channel, along the  $z$ -axis. The face-centered cubic crystal of 5476 Cu atoms were added on the  $x$ -side of Fe slabs (Fig. 1). We applied the periodic boundary condition in all directions. To prevent the Cu infiltration from the opposite side of the narrow channel, the vacuum was introduced right after the Cu slabs in the  $x$ -direction.

Figure 1 shows the initial configuration of the system after the equilibration process at 300 K for 200 ps. We later performed the MD simulations at two different temperatures, i.e., 1360 and 1500 K, which are higher than our obtained melting temperature for the Cu crystal (1357 K). However, these values are lower than the melting point of Fe bulk. In our simulations, a 0 G Pa pressure was applied in all directions.

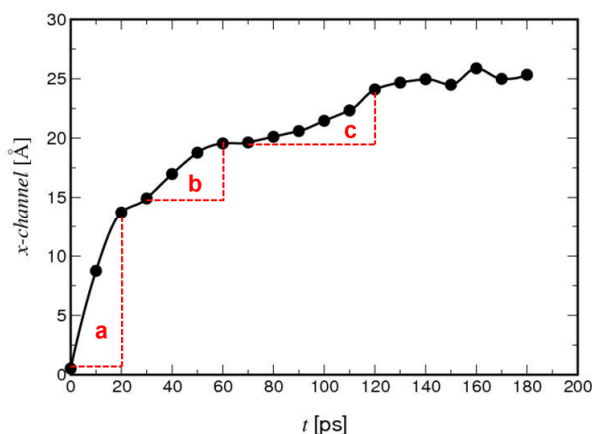
## Results and Discussion

In the brazing process, the system is heated at a temperature higher than the melting point of the filler metal. Cu is one of the most commonly used filler metal to join materials with high melting temperatures, such as Fe. During the brazing process, the Cu liquid infiltrated the narrow channel, or gap, between the two Fe surfaces. The atomic level mechanisms of Cu liquid infiltration through the narrow channel of the Fe bulk can be investigated using MD simulations. In this study, the distance between the two Fe bulks is extremely short, i.e., 20 Å, causing difficulty in experimental observation.

In our study, we measured the position of the Cu liquid front infiltrating the Fe channel every 10 ps. Figure 1 shows the time series of the Cu liquid front in the Fe channel at 1360 K. This temperature is slightly higher than our calculated melting temperature for Cu. From Figure 1, we can deduce that the Cu liquid front moved very rapidly in the beginning until 20 ps. The movement slowed down from 30 ps to 60 ps and considerably slowed down from 70 ps to 120 ps. From this period until the end of the simulation, the positions of the liquid front remained un-changed. As shown in Figure 2, we divided this movement into three time regions, i.e., time region **a**, **b**, and **c**, and the average velocities were calculated as the gradient of the curve in each time region. The velocities in time regions **a**, **b**, and **c** measured 660, 154, and 89 Å/ns, respectively.



**Figure 1. Initial Configuration of the Simulation System. Red and Gray Balls Represent the Fe and Cu atoms, Respectively**

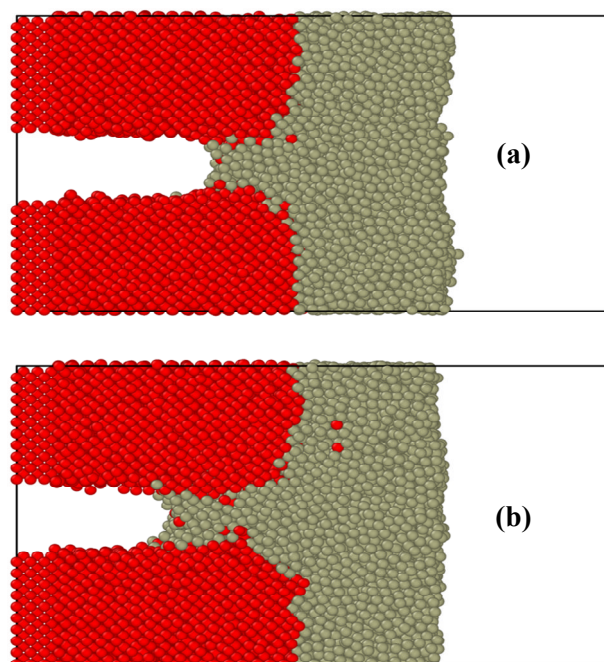


**Figure 2. Time Series of the  $x$ -position of the Front of Cu liquid in the Fe Channel at 1360 K. Different Average Velocities of Liquid Cu Infiltration are Calculated in Three Different Regions, i.e., (a), (b), and (c)**

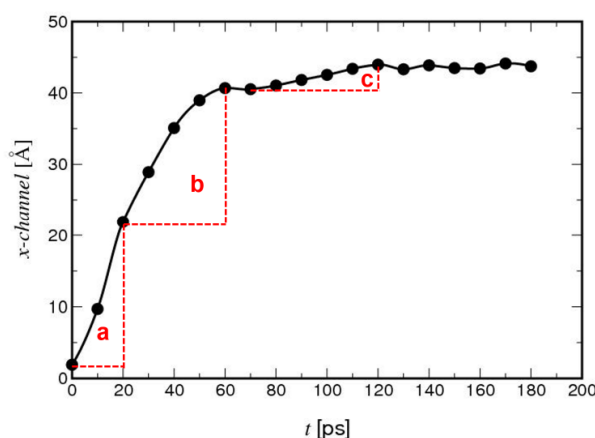
Figure 3 shows the snapshot of the atomic configurations of Cu infiltration through the Fe channel at 30 and 130 ps at  $T = 1360$  K. The figure shows that several Fe atoms on the surface dissolved into the Cu liquid. The concave like surface of the Cu liquid front indicates that the infiltration was preceded by the wetting process of the Fe surfaces by the Cu liquid. The structures of the Fe surfaces were also slightly deformed. This deformation is due to the interaction of the Fe surfaces with the Cu liquid, and the less stable surface structure in comparison with the bulk structure. This process caused narrowing of the channel, and in addition to the viscosity, this factor could possibly cause the decreased infiltration rate of the Cu liquid.

To examine the effect of temperature on the infiltration process, we also investigated the position of the Cu

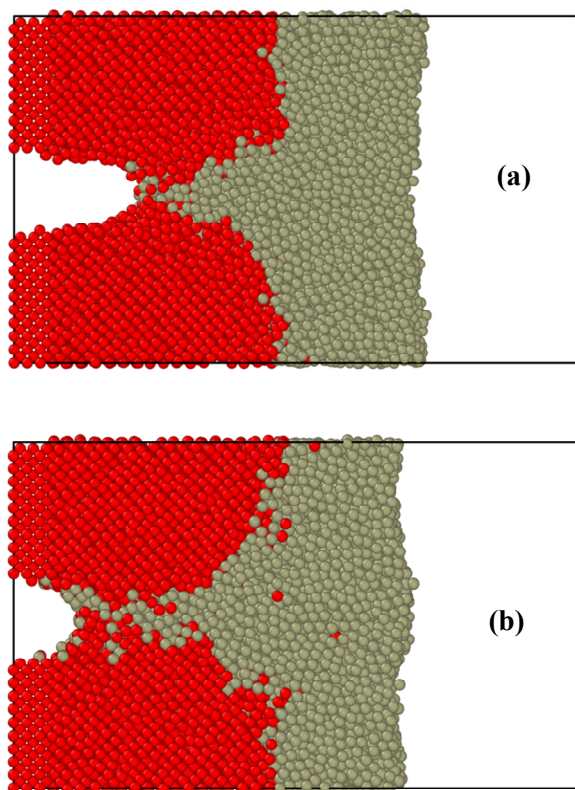
liquid front in the Fe channel  $T = 1500$  K. A similar infiltration mechanism was observed, similar to the one at  $T = 1360$  K. Figure 4 shows that the movement rate of the liquid front significantly decreased at a simulation time of 70 ps. We also divided our results into time region **a**, **b**, and **c** (Figure 4). At  $T = 1500$  K, our calculated velocities in time regions **a**, **b**, and **c** reached 1000, 469, and 69 Å/ns, respectively. The velocities of the front liquids in time region **a** and **b** were higher than that at  $T = 1360$  K. This result is due to the higher energy of Cu atoms at higher temperatures.



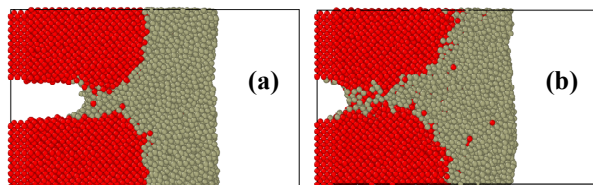
**Figure 3. Snapshot of the Atomic Configurations of Cu Infiltration Through the Fe Channel at  $T = 1360$  K; (a) 30 and (b) 130 ps**



**Figure 4. Time Series of the  $x$ -position of Front Advancement of Liquid Cu in the Fe Channel at 1500 K. Different Average Velocities of Liquid Cu Infiltration were Calculated in Three Different Regions, i.e., (a), (b), and (c)**



**Figure 5. Snapshot of the Atomic Configurations of Cu Infiltration Through the Fe Channel at  $T = 1500$  K; (a) 30 and (b) 130 ps**



**Figure 6. Snapshot of Final Atomic Configurations of Cu Infiltration Through the Fe channel at  $T = 1360$  K (a) and  $T = 1500$  K (b), Respectively**

At time region **c**, the infiltration rate of the Cu liquid front was lower than that at  $T = 1360$  K. Figure 5 shows that at  $T = 1500$  K, the channel was blocked by the Fe atoms from the damaged surfaces. One blockage was observed in the channel (Figure 5 (a)). The number of blockages increased at a simulation time of 130 ps, as shown in Fig. 5 (b).

Figure 6 (a) and 6 (b) show the snapshots of the final configuration of Cu infiltration through the Fe channel at two different temperatures, i.e., 1360 and 1500 K, respectively. At  $T = 1360$  K, the Fe surface was slightly deformed. However, the Cu liquid only filled in a small portion of the channel. At  $T = 1500$  K, the Cu liquid filled almost all the channel, and the Fe surfaces were

highly deformed. From these results, we would suggest further calculations in the future to investigate the tensile strength of each adjoining region.

## Conclusions

We have performed the MD simulation of Fe-joining process using Cu as the filler metal at  $T = 1360$  K and  $T = 1500$  K. We observed in all simulations that wetting of Fe surfaces by the Cu liquid precedes the infiltration process. We also noted that in addition to Cu viscosity, the deformation of the Fe surface caused the decrease in the infiltration rate of the Cu liquid through the Fe channel. At higher temperatures, more surface deformations formed and blocked the liquid flow.

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